



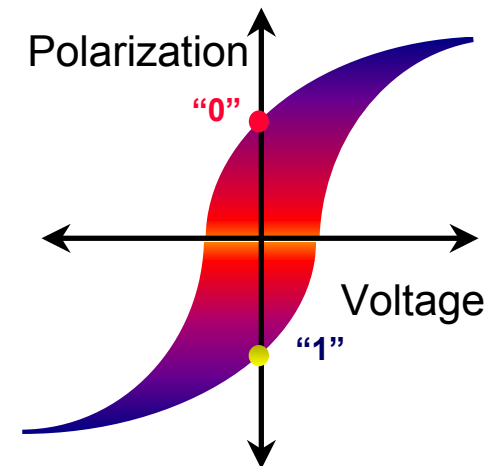
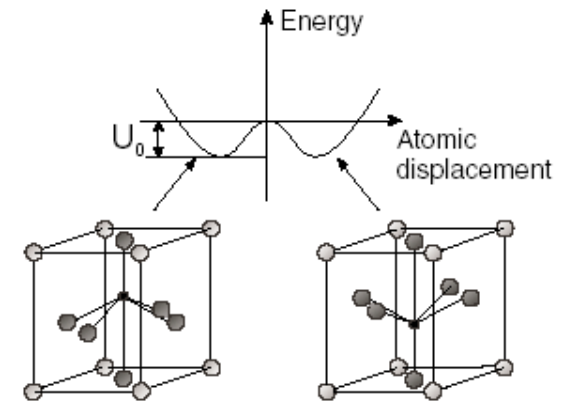
Self-Assembled Polydomain Ferroelectrics and Ferromagnetic Heterostructures

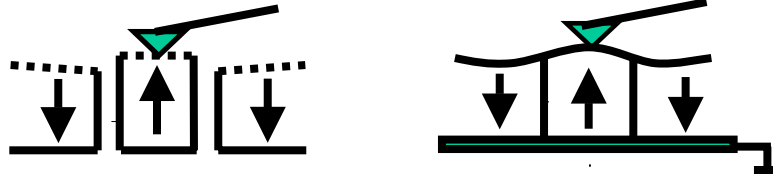
A. L. Roytburd and R. Ramesh, University of Maryland, DMR Award # 0210512

-----Stabilization of Ferroelectric Domains after Switching-----

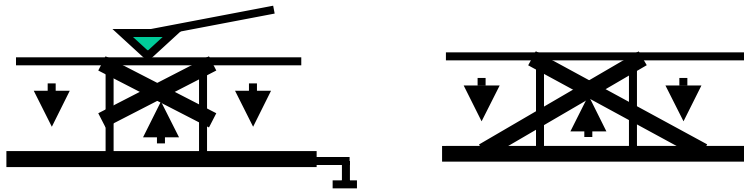
An important example of “functional oxides microelectronics” is ferroelectric(FE) thin films grown on metal or oxide electrode layers for use in nonvolatile memories: Ferro-Electric Random Access Memory(FERAM) which employs the natural characteristics of FEs to be in one of two polarization states corresponding to “1” or “0”. The density of the storage information now is up to 28G bit/cm² and densities up to 150 G bit/cm² have been extrapolated from sizes of individual domains [T. Tybell *et al*, *Phys. Rev. Lett.* **89**, 097601 (2002)].

However, it is often observed that FE films used for FERAM exhibit significant reversal of the domain structure as a result of partial and complete back switching. Therefore, the stability of local polarization reversal remains to be an important issue to fabricate reliable memory devices of high density.

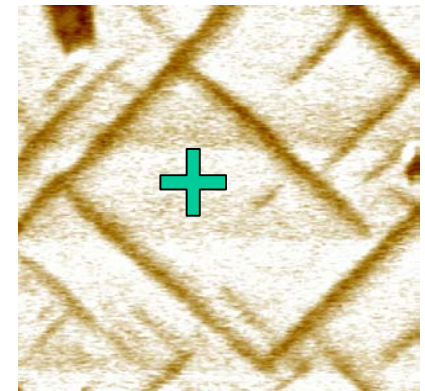
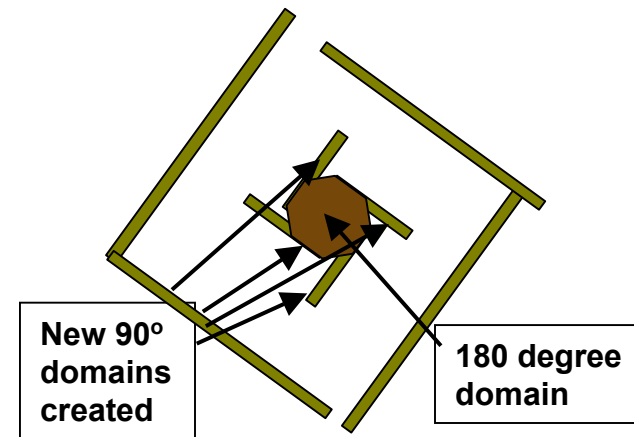




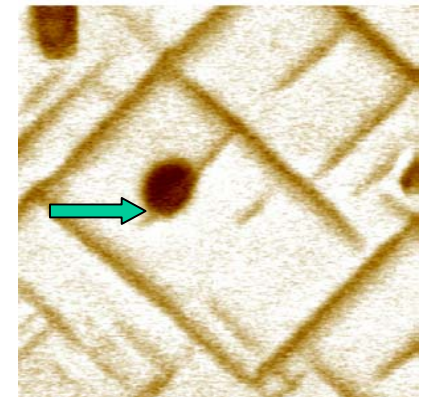
The local electric field creates a 180° domain and induces stress due to the opposite signs of piezostress inside and outside of the domain.



The piezostress relaxes by the formation of 90° domains.
After the electric field is off, 90° domains form a stable structure.



Before(up) and after(down) writing



It has been demonstrated that a cylindrical 180° domain can be stabilized by the formation of 90° domains to relax stress that arises due to the opposite signs of the converse piezoelectric effects in the switched 180° domain and the unswitched film surrounding. The formation of 90° domains is a possible mechanism for the polarization stabilization in FE films.

See: L. Chen, J. Ouyang, C. S. Ganpule, V. Nagarajan, R. Ramesh, and A. L. Roytburd, *Appl. Phys. Lett.* 84, 254(2004).



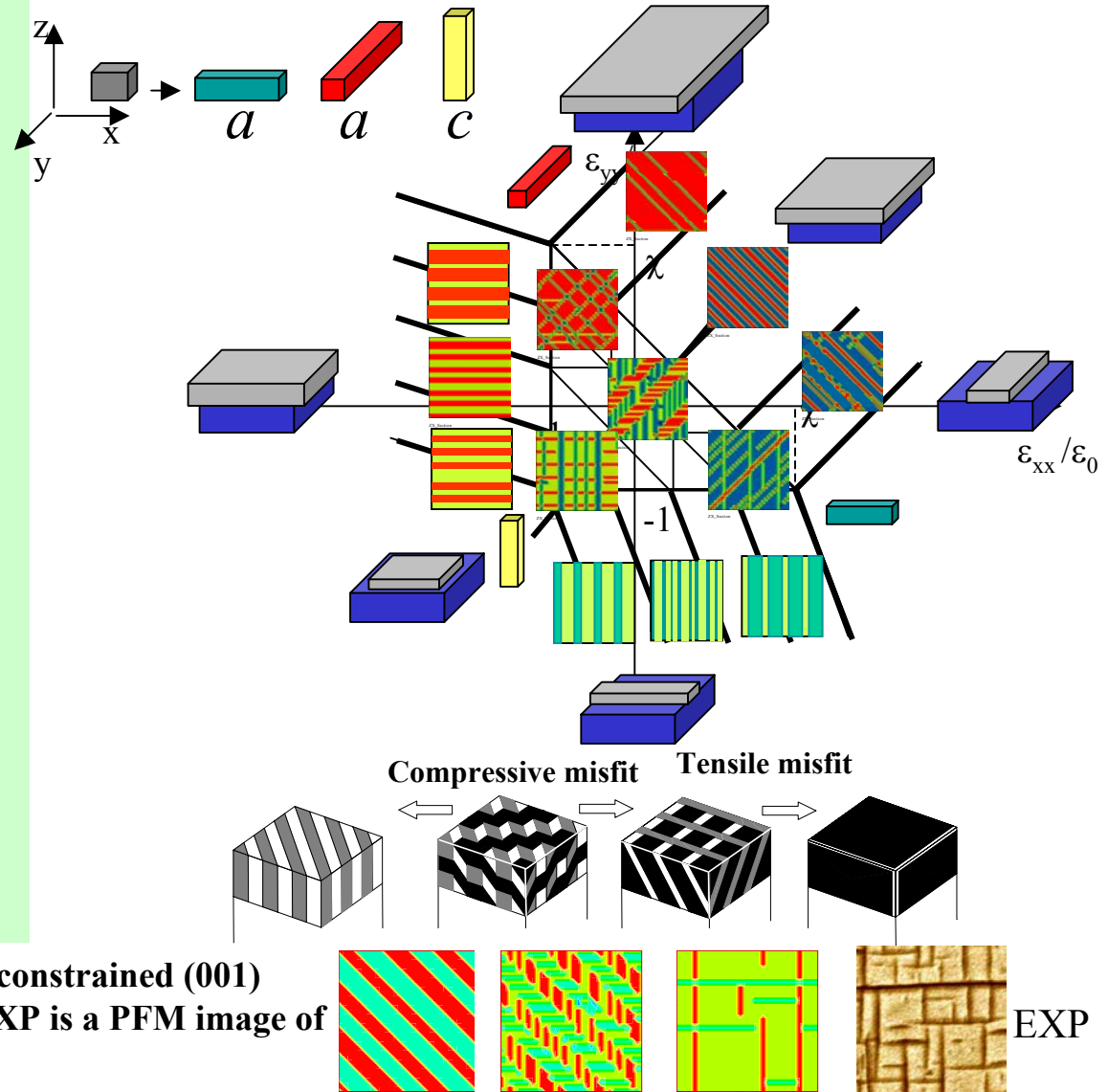
Self-Assembled Polydomain Ferroelectrics and Ferromagnetic Heterostructures

A. L. Roytburd and R. Ramesh, University of Maryland, DMR Award # 0210512

----- Engineering Polydomain Structures in Constrained Ferroelectric Films -----

Polydomain structures containing different types of domains (a , a and c) arising as a result of a ferroelectric phase transformation in a constrained film.

Phase-field modeling demonstrates that by manipulating constraint parameters, such as misfit, elastic properties of substrates and thickness of the film, it is possible to **engineer different polydomain architecture**. The polydomain structures consisting of a - a domains are strong dielectrics. The polydomain structures consisting of a - c domains are potentially giant piezoelectrics.



Figures shows polydomain structures in a constrained (001) layers at different biaxial misfit, ($\epsilon_{xx}, \epsilon_{yy}$). EXP is a PFM image of a PZT film on SrTiO₃ substrate.



Self-Assembled Polydomain Ferroelectrics and Ferromagnetic Heterostructures

A. L. Roytburd and R. Ramesh, University of Maryland, DMR Award # 0210512

----- Engineering Polydomain Structures in Constrained Ferroelectric Films -----

Publications

1. J.Slutsker, A.Artemev and A.L.Roytburd, "Morphological transitions of elastic domain structures in constrained layers", *Journal of Applied Physics* **91**, 9049-9058, (2002)
2. J.Slutsker, A.Artemev and A.L.Roytburd, "Elastic domain architectures in constrained ferroelectric films", in *Fundamental Physics of Ferroelectrics*, (ed.R.E.Cohen), *AIP Conference Proceedings*, pp.266-276, (2002)
3. J.Slutsker, A.Artemev and A.L. Roytburd "Modeling of martensitic transformation in adaptive composites", *J.Phys.IV* **112**, 115-118 (2003)
4. J.Slutsker, A.Artemev and A.L.Roytburd, "Engineering of elastic domain structures in a constrained layer", *Acta Mater.*, 52, 1731-1742 (2004)

Invited talks

1. J.Slutsker and A.L.Roytburd: "Theory and Modeling of Martensitic Transformations in Films and Composites", *ICOMAT'02*, Helsinki, Finland, June 2002
2. J.Slutsker and A.L.Roytburd, "Engineering of Domain Structures in Films and Composites", TMS Annual Meeting, San Diego, California, March 2003
3. A.L.Roytburd, "Nano- and Micro Elastic Domains in Constrained Ferroelastic Layers" TMS 2003, San Diego, CA, March, 2003
4. A.L.Roytburd, J.Slutsker and A.Artemev " Modeling of Composites with Self-Assembling Micro- and Nano-Structures ",TMS 2004, Charlotte, NC, March, 2004
5. A.L.Roytburd, "From Martensite to Self-Assembled Nanostructures", TMS 2004, Charlotte, NC, 2004.



Self-Assembled Polydomain Ferroelectrics and Ferromagnetic Heterostructures

A. L. Roytburd and R. Ramesh, University of Maryland, DMR Award # 0210512

-----Enhancement of Piezoelectric Response in Nanoscale Heterostructures-----

Enhanced piezoresponse of ferroelectric films can be obtained by reducing substrate clamping. For polydomain films, decrease of clamping can lead to giant piezoresponse due to 90 degree domain walls movement.

Epitaxial ferroelectric films were patterned to nanosize islands via focused ion beam (FIB) milling (Fig. 1). For $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ films grown on the Si and SrTiO_3 (STO) substrate, effects of size and geometry (square vs. rectangular with different lateral aspect ratio) of islands on their domain structures and properties have been investigated.

In the square shape islands the maximum longitudinal piezoelectric constant d_{33} reaches ~ 250 pm/V on STO substrate (Fig. 2) and ~ 400 pm/V on Si substrate, which is about 3-4 times the theoretical value of 87 pm/V for a single domain crystal.

Publication: V. NAGARAJAN, A. ROYTBURD, A. STANISHEVSKY, S. PRASERTCHOUNG, T. ZHAO, L. CHEN, J. MELNGAILIS, O. AUCIELLO AND R. RAMESH, "Dynamics of ferroelastic domains in ferroelectric thin films", *Nature Materials*, Vol.2, p.43(2003)

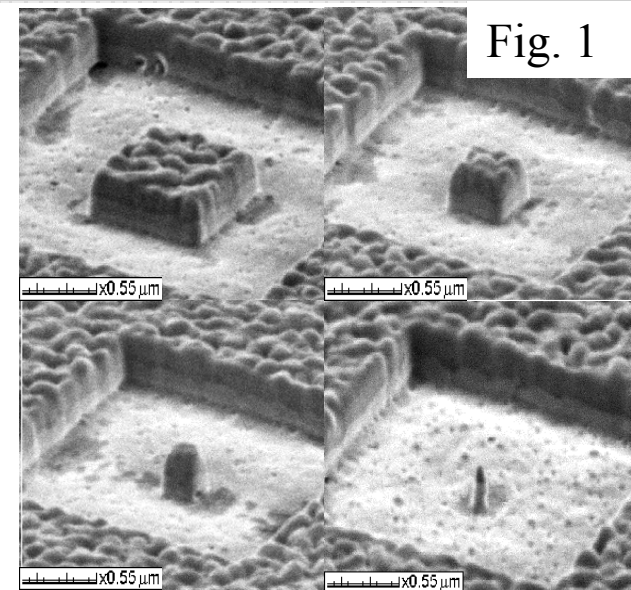


Fig. 1

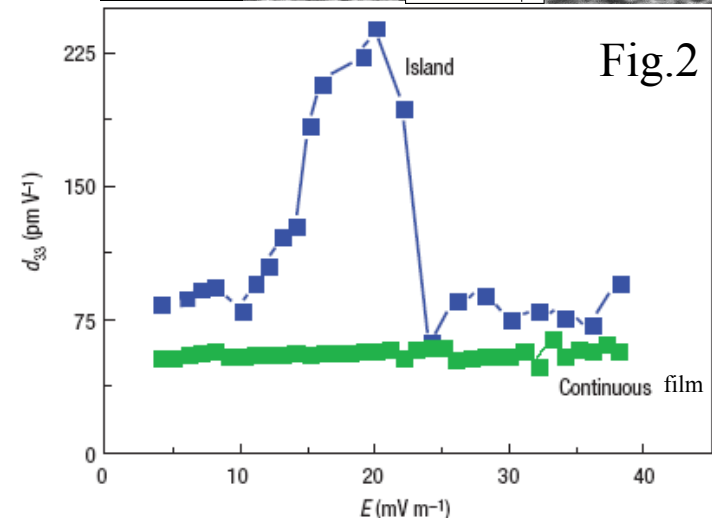


Fig.2



Self-Assembled Polydomain Ferroelectrics and Ferromagnetic Heterostructures

A. L. Roytburd and R. Ramesh, University of Maryland, DMR Award # 0210512

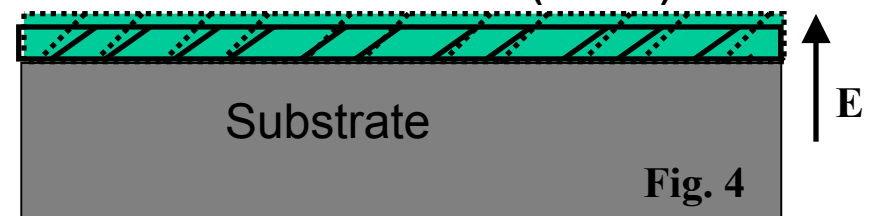
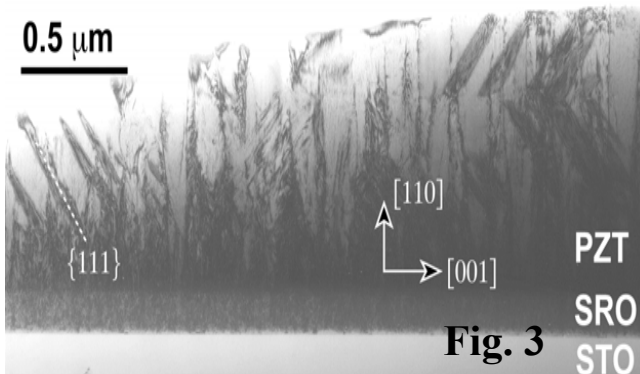
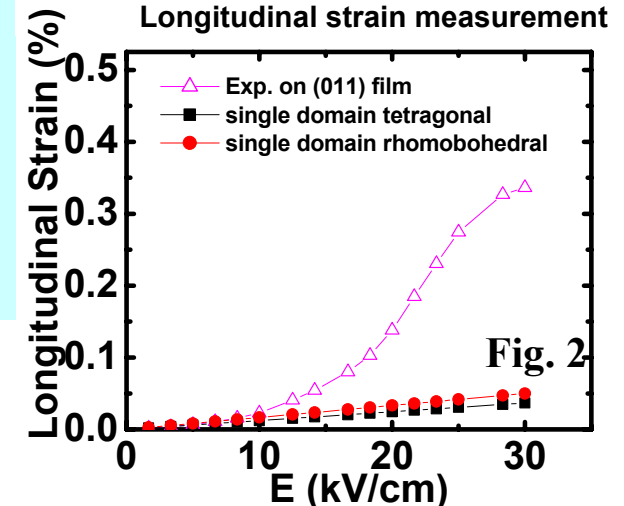
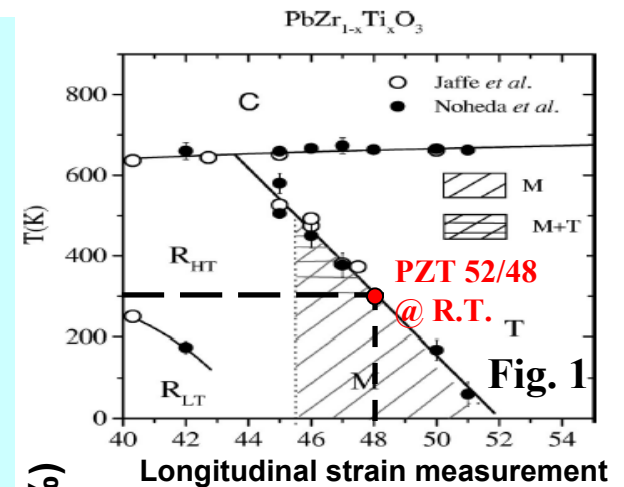
--- Ultrahigh Piezoelectric Response in a Ferroelectric $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$ Thin Film---

➤ Lead zirconate titanate ferroelectric solid solutions- $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$ have excellent piezoelectric properties, especially in its composition range near the morphotropic phase boundary ($x \sim 0.5$) (Fig. 1).

➤ The intrinsic piezoelectric constant for a clamped PZT 52/48 film is calculated to be in the order of 100 pm/V. By experimental studies of epitaxial films in different orientations, a large piezoelectric response has been found in the (011) oriented PZT 52/48 film. The effective piezoelectric constant is ~ 1000 pm/V, **an order of magnitude higher** than the intrinsic value. It may have important potential applications in next generation MEMS devices. (Fig. 2)

➤ The TEM cross sectional picture showed a certain domain structure existing in the (011) oriented PZT 52/48 film. (Fig. 3)

➤ It is suggested that the high piezoelectric response can be understood on the basis of a highly mobile polydomain structure of the film under investigation. (Fig. 4)





Self-Assembled Polydomain Ferroelectrics and Ferromagnetic Heterostructures

A. L. Roytburd and R. Ramesh, University of Maryland, DMR Award # 0210512

Education:

- Lang Chen, Graduate student, Supported by this grant.
- Jun Ouyang, Graduate student, Supported by this grant.
- Zhengkun Ma, Graduate student.
- Jianhua Li, Graduate student.

Collaborators:

- Dr. J. Slutsker, Department of Materials Science and Engineering, University of Maryland, College Park and Materials Science and Engineering Lab, NIST.
- Professor J. Melngailis, Department of Electrical Engineering, University of Maryland, College Park
- Dr. V. Nagarajan, Institute for Electronic Materials, IFF, Forschungszentrum, Julich, D-42425, Germany
- Professor A. Artemev, Carleton University, Canada.